

Bench and Pilot-Scale Characterization of Low Temperature Selective Catalytic Reduction of NO_x

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Summary

The objective of this work is to develop a strategy for low temperature NO and SO_x control. To achieve this control, dry sorbent and ammonia are to be injected into coal boiler flue gas upstream of a conventional low temperature baghouse with catalyzed monoliths placed within the filter bags. To this end, the present study has characterized the activity and selectivity of a commercial low temperature NO reduction catalyst using ammonia as the reductant. Tests were performed in a bench-scale, fixed-bed reactor using monolith supported catalysts to cover a range of feedstream conditions that are representative of industrial-scale coal-fired boilers. Synthetic flue gas mixtures at temperatures of 160 to 350_C were passed through a catalyzed monolith while the inlet and exit gas compositions were quantitatively analyzed using an FTIR spectrometer, which allowed both the activity and selectivity of the catalyst to be monitored. The measurements show that activity peaks at nearly 90% NO conversion between 240 and 300_C, while selectivity (to N₂) peaks at nearly 98% between 150 and 200_C. Similar results were obtained when the catalyst was tested on a pilot-scale combustor (0.5 MMBtu/h) fired on natural gas.

Bench-Scale Testing

The goals of the bench-scale tests were to identify a NO reduction catalyst compatible with the typical operating conditions for an industrial boiler and establish the range of effective operating conditions of the catalyst in terms of conversion efficiency and selectivity. With this detailed information, a design for testing at the demonstration-scale could be developed. To this end, we examined the effectiveness of a low-temperature ammonia selective catalytic reduction (SCR) catalyst supplied by Engelhard Corp., commercial name NOxCATtm 920 LT. This is a precious metal catalyst and exhibits two typical features of precious metal catalysts: good low temperature activity; and a tendency to produce NO as a side product during the reduction of NO.

Cylindrical monolith cores were tested over a range of inlet conditions by varying feedstream composition, space velocity and gas temperature. Sensitivity of the catalyst to the presence of SO₂ was pronounced at low temperatures but less significant at higher temperatures. Optimal conversion and

product composition occurred when the ammonia was stoichiometrically matched to the NO concentration. Conversion of NO was affected by the presence of water vapor, but selectivity of the reaction to N_2 was not. Decreasing space velocity increased NO conversion modestly, but greatly enhanced selectivity to N_2 . At lower feedstream temperatures, selectivity is enhanced and conversion falls off. At higher temperatures, conversion falls off because an increasing amount of the ammonia reductant is directly oxidized to NO.

The observations of the NOxCATtm 920LT regarding conversion and selectivity can be explained by postulating two types of adsorption sites, where both are active for conversion of NO, but one type of site (type-I) produces N_2 and the other (type-II) produces N_2O . If the type II site is readily poisoned by the presence of SO_2 or H_2O , then an improvement in selectivity and a decrease in overall conversion would be observed as is the case in our results. The type I site is apparently insensitive or at least far less sensitive to these poisoning species.

Preliminary PilotScale Testing

To verify the observations from the bench-scale tests, pilot-scale tests have been performed using the Down-Fired Combustor (DFC) located in the Energy and Fuels Research Center at Penn State. This 0.5 MMBtu/h facility can be fired on natural gas, coal, coal water slurry or combinations of fuels. Preliminary tests firing natural gas were performed to examine the NO_x reduction capability of the catalyst in the absence of appreciable SO_2 , but in a practical flue gas. A cylindrical monolith was obtained from Engelhard for testing, and was mounted in ductwork attached to the exhaust system of the DFC. External heaters provided temperature control over the flue gas entering the catalyst. However, over the range of inlet temperatures considered, the NO_x concentration supplied to the catalyst varied by roughly 50 ppm. Two space velocities were considered. The pilot-scale results were similar to the bench-scale results, confirming the basic trend that selectivity and (to a lesser extent) conversion increase with decreasing space velocity.

Future Work

Additional pilot-scale studies firing on coal and using injection of both sodium bicarbonate and ammonia to obtain combined SO_x and NO_x control will be performed in May and June, 1997. From the bench and pilot-scale results and design for testing on the demonstration-scale will be developed and implemented in 1998 if tests warrant continued pursuit of this strategy.

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